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Random laser (RL) emission in Nd^{3+} doped potassium gadolinium tungstate—$KGd(WO_4)_2:Nd^{3+}$—crystal powder is demonstrated. The powder was excited at 813 nm in resonance with the Nd^{3+} transition $^4I_{9/2} \rightarrow {}^4F_{5/2}$. RL emission at 1067 nm due to the $^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition was observed and characterized. An intensity threshold dependent on the laser spot area and bandwidth narrowing from ≈2.20 nm to ≈0.40 nm were observed and measured. For a beam spot area of 0.4 mm^2, a RL threshold of 6.5 mJ/mm^2 (90 MW/cm^2) was determined. For excitation intensity smaller than the RL threshold, only spontaneous emission from level $^4F_{3/2}$ with decay time in the tens microsecond range was observed, but for excitation above the RL threshold, significant shortening of excited level lifetime, characteristic of a stimulated process was found. The overall characteristics measured show that $KGd(WO_4)_2:Nd^{3+}$ is an efficient material for operation of solid state RLs in the near-infrared. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4913390]

I. INTRODUCTION

Random lasers (RLs) are sources based on the scattering induced stimulated emission of light in disordered gain media.[1–3] Two kinds of random media are exploited to operate RLs. In one case, the active medium is exemplified by a laser-dye containing dielectric or metal particles as scatterers;[1,4–7] in another case, a powder consisting of grains that act as amplifying medium and scatterers are used. In this second case, the particles used are semiconductors or dielectric crystals doped with rare-earth ions.[2,8–10]

One of the mostly studied RL systems is based on powders of neodymium (Nd^{3+}) doped microcrystals because of the low threshold and high efficiency of the ≈1.06 μm emission associated to the Nd^{3+} transition $^4F_{3/2} \rightarrow {}^4I_{11/2}$.[2] Besides the near-infrared RL emission, ultraviolet RL emission was also demonstrated due to an orange-to-ultraviolet frequency upconversion process[11] and by electrical pumping.[12]

Currently, identification of new materials for efficient RL operation is of great interest. Materials used for conventional lasers are natural candidates and, in this sense, we suggest that the potassium gadolinium tungstate laser crystal—$KGd(WO_4)_2:Nd^{3+}$—is a good material, since it possesses excellent optical, thermal, and mechanical properties[13] for being used in conventional lasers operating at 1067 nm. In addition, the strong Raman line at 1180 nm can be used for self-frequency conversion.[14] Besides its large damage threshold (10^{11} W/cm^2) and large linear refractive index (≈2) reported in Ref. 13, $KGd(WO_4)_2:Nd^{3+}$ can be doped with high Nd^{3+} density (up to 8 mol % with respect to the Gd site) without any fluorescence quenching and crystal quality deterioration.[14] The absorption and stimulated emission cross sections of Nd^{3+} in $KGd(WO_4)_2$ are also larger than in other laser host crystals due to its large refractive index.

In the present paper, we report RL action in a $KGd(WO_4)_2$: Nd^{3+} crystal powder. The emitted wavelength at 1067 nm was obtained by exciting the medium at 813 nm, in resonance with the Nd^{3+} transition $^4I_{9/2} \rightarrow {}^4F_{5/2}$. The RL intensity, spectral bandwidth, and temporal dynamics dependence with the excitation pulse energy (EPE) were characterized. Also, the dependence of the RL threshold on the excited spot area was studied.

II. EXPERIMENTAL DETAILS

A potassium gadolinium tungstate laser crystal—$KGd(WO_4)_2:Nd^{3+}$—(labeled as KGW:Nd), with Nd^{3+} concentration of 4 mol % (INCROM Ltd., Russia), was crushed and the grains passed through calibrated sieves to obtain a powder. The size distribution of the grains with average dimensions of ≈5 μm was determined using a scanning electron microscope. The powder was excited by an optical parametric oscillator pumped by a Q-switched Nd:YAG laser (7 ns, 10 Hz), focused on the sample by a 10 cm biconvex lens. The pump wavelength 813 nm was chosen to optimize the fluorescence signal at 1067 nm. The EPE was controlled by a pair of polarizers and measured with a calibrated silicon photodiode. The angle between the normal to the sample and the incident beam was 10° to favor scattered light collection from the front face of the sample using a 5 cm focal length

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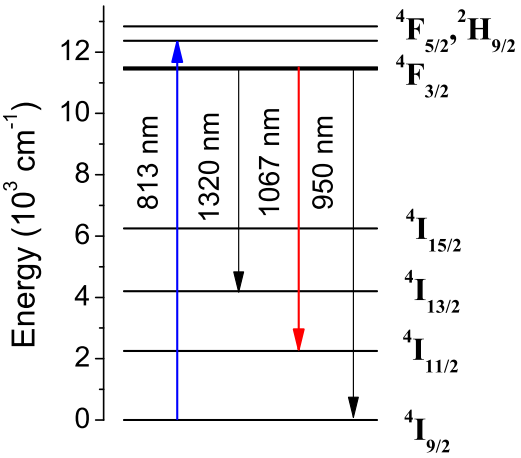

FIG. 1. Energy level diagram including the relevant Nd^{3+} transitions. The incident light at 813 nm promotes electrons to the $^4F_{3/2}$ level and from there occur the main radiative transitions with the wavelengths indicated in the figure.

lens. The collected light was focused by a 20 cm lens at the entrance of a spectrometer (resolution: 0.1 nm) equipped with a cooled CCD. The temporal behavior of the emitted light was characterized using a photodetector with a nanosecond response. A long-pass filter with transmission higher than 80% for wavelengths above 850 nm was inserted before the entrance slit of the spectrometer to block the elastically scattered laser light.

III. RESULTS AND DISCUSSION

Figure 1 represents the Nd^{3+} energy levels and the laser induced transitions of interest for the present experiments. By exciting the KGW:Nd powder at 813 nm, the electrons undergo transitions from the Nd^{3+} ground state $^4I_{9/2}$ to the excited state $^4F_{5/2}$. From there, the Nd^{3+} ions decay to lower energy states with infrared emissions corresponding to transitions $^4F_{3/2}\rightarrow{}^4I_{13/2}$ (1320 nm), $^4F_{3/2}\rightarrow{}^4I_{11/2}$ (1067 nm), and $^4F_{3/2}\rightarrow{}^4I_{9/2}$ (950 nm). Our work focuses on the well-known laser transition $^4F_{3/2}\rightarrow{}^4I_{11/2}$.[2]

Figure 2 presents the spectra corresponding to the transition $^4F_{3/2}\rightarrow{}^4I_{11/2}$ (1067 nm), when the KGW:Nd powder is

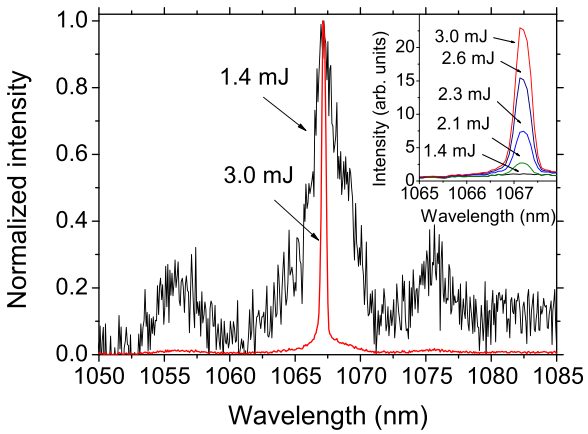

FIG. 2. Normalized spectra of the transition $^4F_{3/2}\rightarrow{}^4I_{11/2}$ obtained by pumping the system at 813 nm (pump spot size: 2.5×10^{-3} cm^2) with EPE smaller (1.4 mJ) and larger (3.0 mJ) than the RL threshold. The inset illustrates the behavior of the 1067 nm band when the EPE is changed.

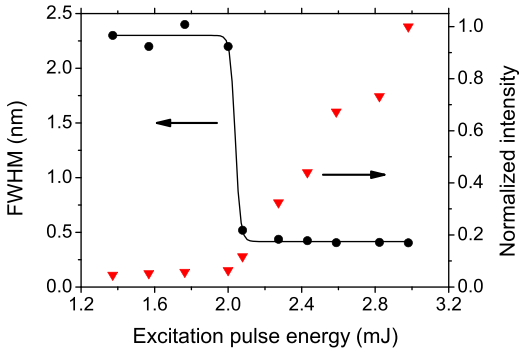

FIG. 3. Full width at half maximum and normalized intensity of the band centered at 1067 nm as a function of the excitation pulse energy. The pump spot size was 2.5×10^{-3} cm^2. The curve shown is a guide to the eyes.

excited at 1.4 mJ and 3.0 mJ. The weak bands centered at ≈1057 nm and ≈1075 nm are Stark-level satellites. For completeness, the inset of Fig. 2 illustrates the behavior of the 1067 nm emission, as the EPE is varied.

Figure 3 shows the full width at half maximum (FWHM) of the band centered at 1067 nm versus the EPE. A narrowing from 2.25 to 0.40 nm is observed for EPE larger than 2.0 mJ. Notice also from Fig. 3 that when the EPE is increased beyond ≈2.0 mJ, a large increase in the emitted intensity is obtained. The intensity grows by a factor of ≈20 when the EPE is increased from 1.9 mJ to ≈3 mJ. These results indicate a RL threshold at EPE ≈ 2.0 mJ.

The RL behavior was also investigated with respect to the dynamics of the 1067 nm emission. Figure 4 shows the time decay of the generated light after excitation with the 7 ns laser pulse duration. The signal detected gives information about the lifetime of the state $^4F_{3/2}$. The inset in Fig. 4 shows the signal decay for excitation at 1.9 mJ. The experimental data show a temporal behavior that can be described by a double-exponential curve $[Ae^{-t/\tau_1}+Be^{-t/\tau_2}]$, with $\tau_1=12\,\mu$s and $\tau_2=85\,\mu$s. The double-exponential decay is attributed to the Nd^{3+}-Nd^{3+} interaction that is expected for a large Nd^{3+} concentration (4%) present in the original crystal, on the basis

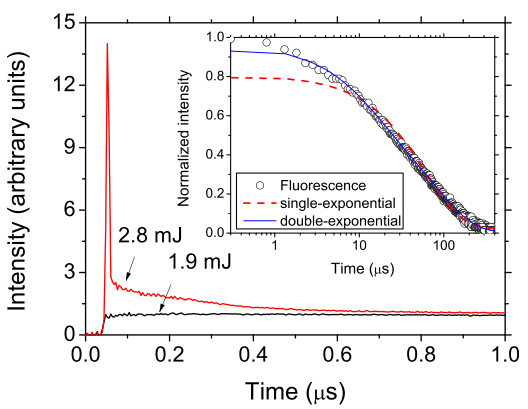

FIG. 4. Time evolution of the fluorescence from the $^4F_{3/2}$ level for EPE smaller (1.9 mJ) and larger (2.8 mJ) than the random laser threshold corresponding to an excited spot area of 2.5×10^{-3} cm^2. The inset shows the signal corresponding to 1.9 mJ. Notice that the data is better fitted with a double-exponential.

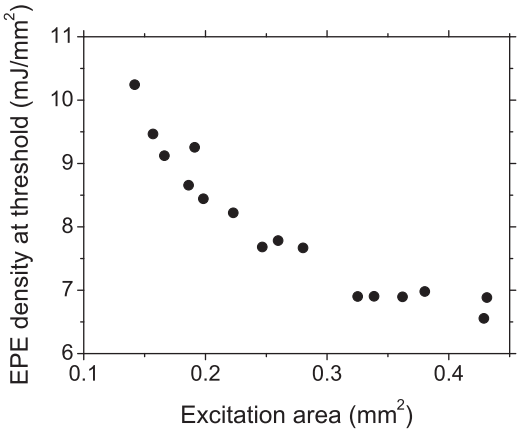

FIG. 5. Excitation pulse energy density threshold as a function of the illuminated area.

of results from other Nd^{3+} doped crystals.[15,16] On the other hand, for EPE larger than 2.0 mJ, we observed a fast decay (≈7 ns) of the signal followed by a slow decay in the microsecond range. The fast component is attributed to the stimulated emission, while the slow decay is due to the spontaneous decay of excited Nd^{3+} that were not influenced by stimulated emission.

The results presented in Figs. 3 and 4 were obtained by focusing the laser beam into a spot area of 0.25 mm^2 corresponding to EPE of 2.0 mJ (power density: 110 MW/cm^2). However, as was already shown,[2,17] the intensity threshold, or equivalently the EPE density threshold, is dependent on the directly excited area/volume in the sample. In the present study, this behavior was observed measuring the EPE density threshold for various excitation spot areas by changing the position of the focusing lens along the pump beam direction. Figure 5 shows the EPE density threshold versus the excitation area. This behavior is due to a balance between the volume effectively excited and the pathways travelled by the incident and the emitted photons inside the gain volume.[2,17] Small pump volumes require high EPE density to obtain RL emission, because the time interval elapsed by photons inside the gain volume is small. The measurements were not extended for larger areas because of the limited maximum output of the excitation laser available. A comparison between the present results with the ones for $NdAl_3(BO_3)_4$ reported in Ref. 18, which is an efficient material for RL, can be made considering the data of Fig. 5, where an EPE density of ≈6.5 mJ/mm^2 (90 MW/cm^2) was observed for a spot area of 0.4 mm^2. This result is of the same order of magnitude as the one for $NdAl_3(BO_3)_4$ for equal exciting spot area.

IV. SUMMARY

In summary, RL emission in a KGW:Nd crystal powder at 1067 nm was demonstrated for the first time, by exciting the system at 813 nm in resonance with the Nd^{3+} transition $^4I_{9/2} \rightarrow {}^4F_{5/2}$. The RL nature of the emitted photoluminescence was characterized by bandwidth narrowing, linear growth of the emitted intensity at a large slope above a threshold, and reduction of the signal time decay from several microseconds to ≈7 ns, when the RL regime is reached. The EPE density threshold for a pump spot area of 0.25 mm^2 was 110 MW/cm^2, and decreased ≈40% when the spot area increased from 0.14 mm^2 to 0.4 mm^2.

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